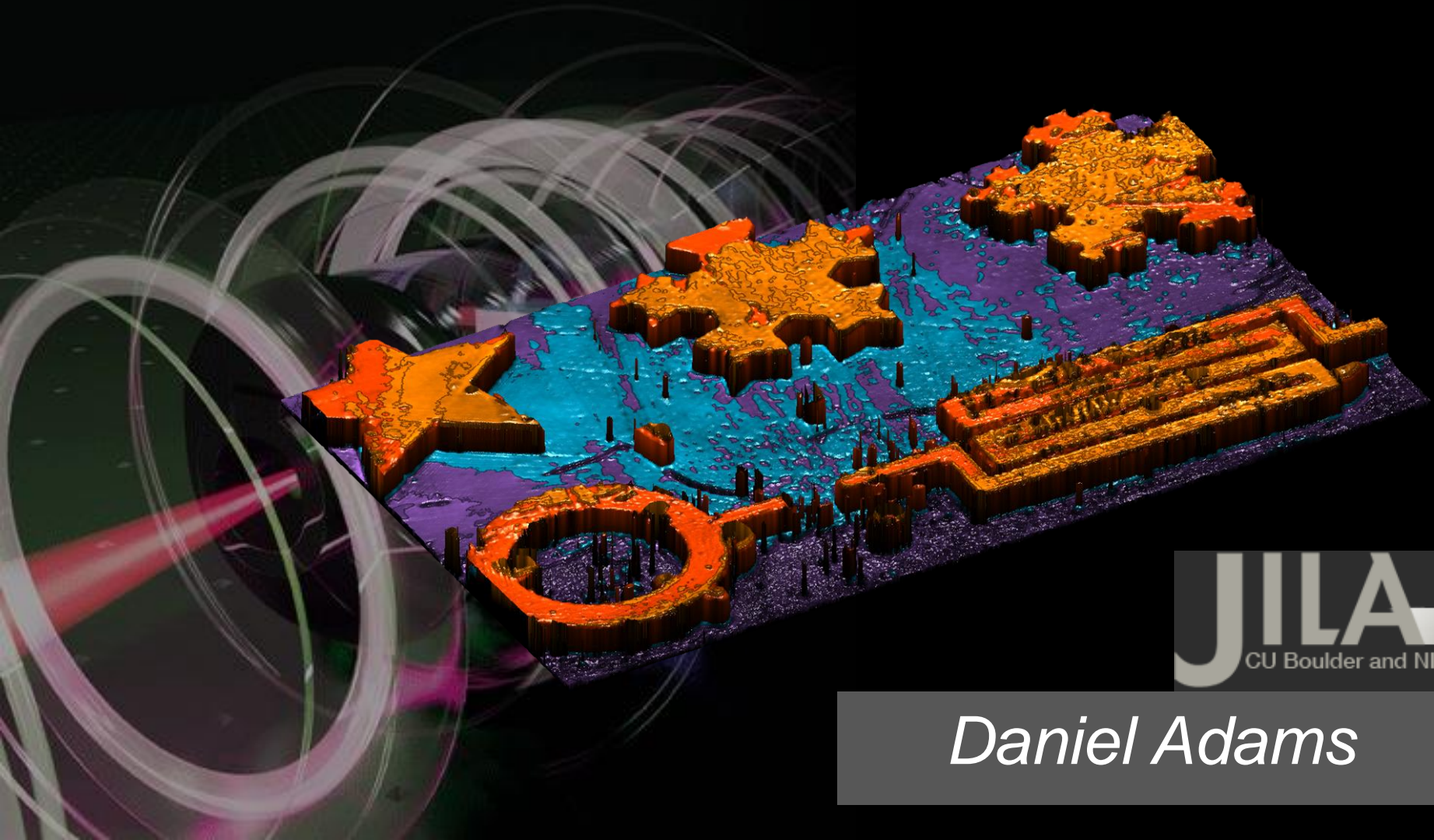
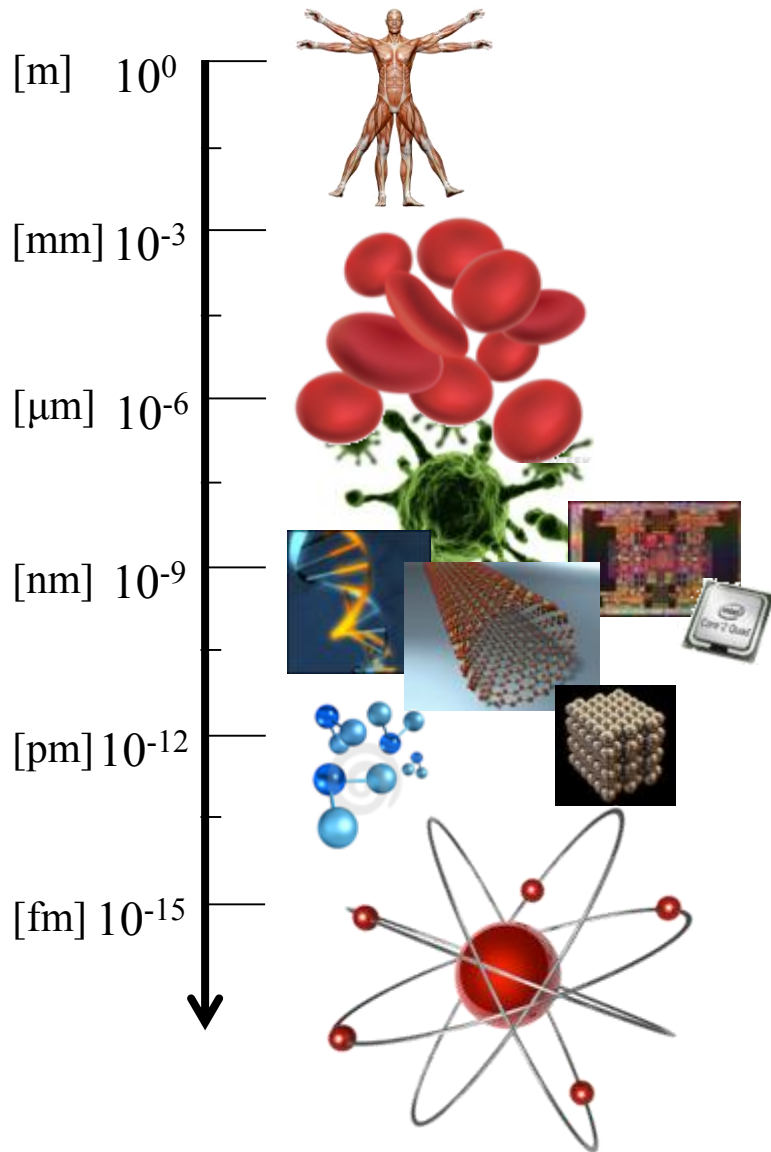


# Tabletop-Scale EUV Coherent Phase-And-Amplitude Imaging Using High Harmonics



*Daniel Adams*

# Coherent EUV Nano-probe



Human

Blood Cell

Virus

Transistor Nanotubes

DNA

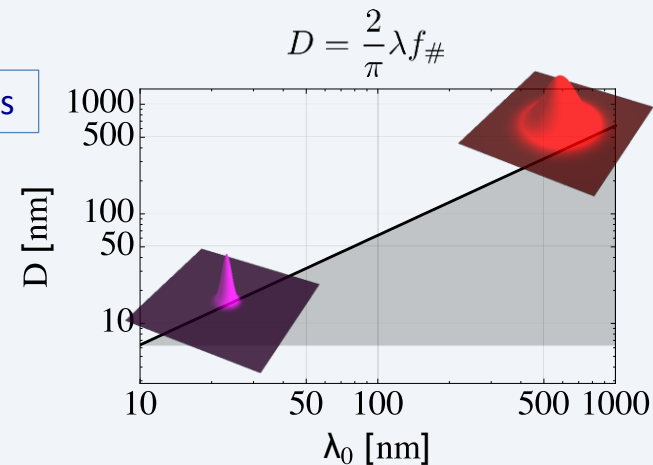
Molecular  
Atomic Spacing

Nuclear  
Diameter

## Uncertainty Principle

(Gaussian,  $e^{-2}$  Intensity)

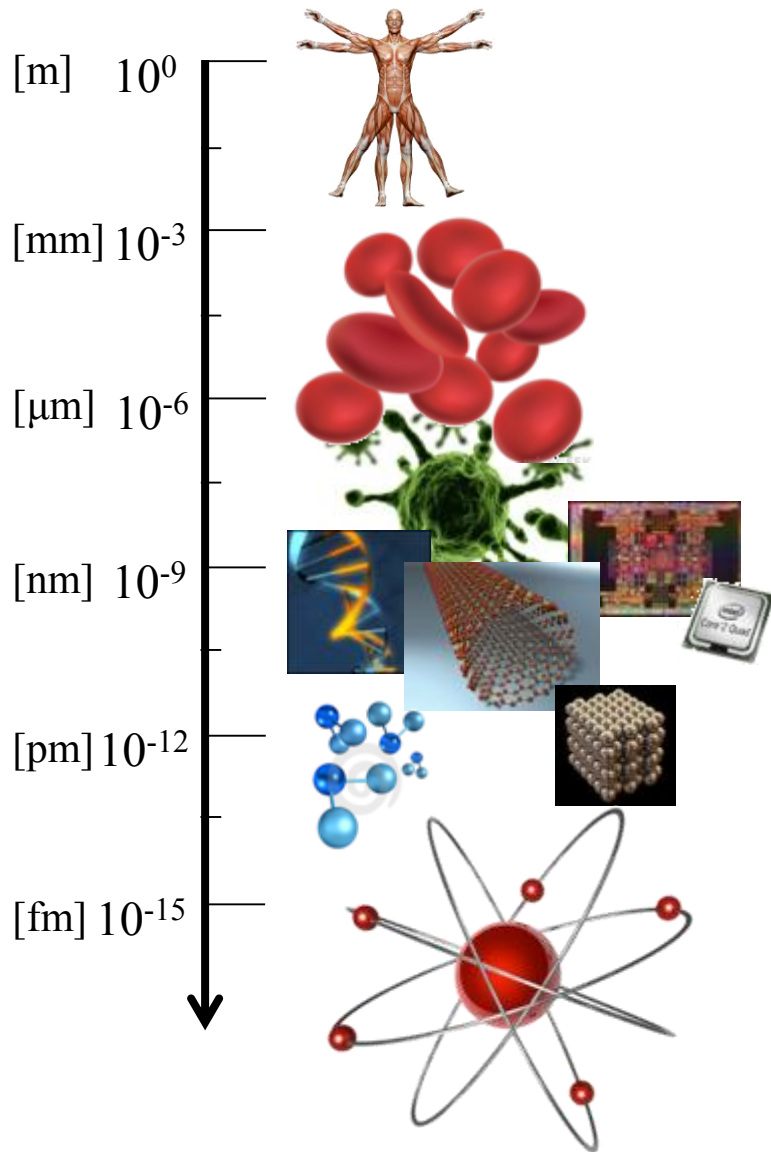
$$\Delta x \Delta k \sim 1.2$$



**Coherent X-rays are ideal probes of the nanoworld:**

- Penetrate thick objects
- Elemental and chemical specificity
- Image small features in 3D at wavelength limit
- Capture all dynamics relevant to function

# Coherent EUV Nano-probe



Human

Blood Cell

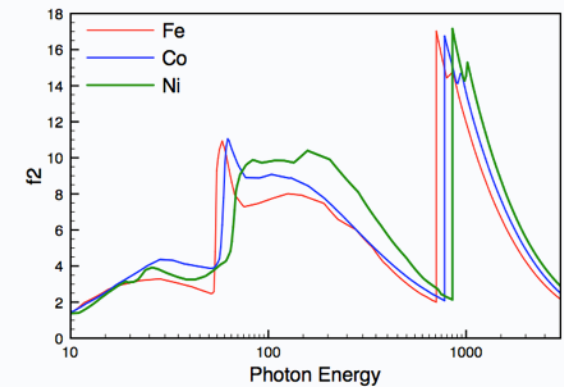
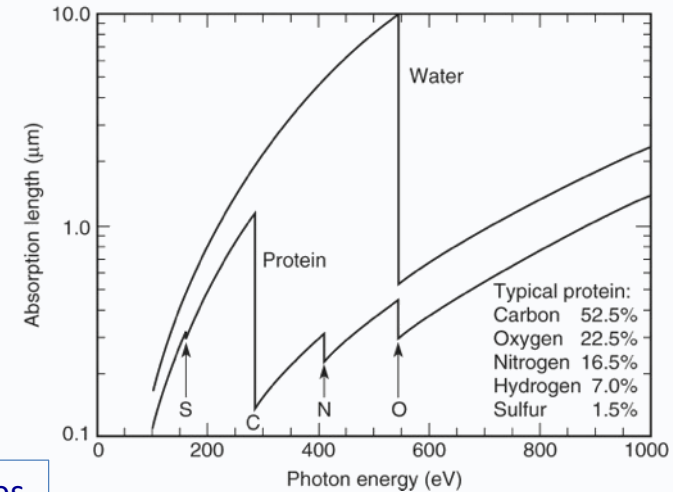
Virus

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# Coherent EUV Sources

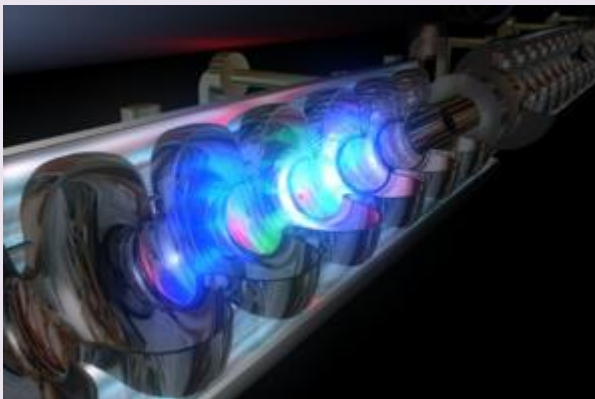
## FACILITY SCALE SOURCES

### Synchrotron



Miao et al., *Nature*, **400**, 342 (1999)

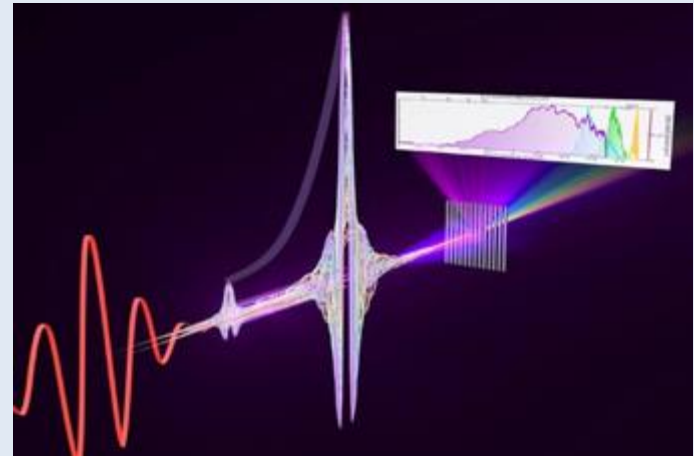
### Free Electron Laser



Chapman et al., *Nature Phys.* **2**, 839 (2006)

## TABLETOP SOURCES

### High harmonics



T. Popmintchev et. al., *Science*, **336**, 6086, (2012)

### EUV laser source at 13nm




Martz et al, *Optics Letters*, **35**, 1632 (2010)



# Coherent EUV Sources

## FACILITY SCALE SOURCES

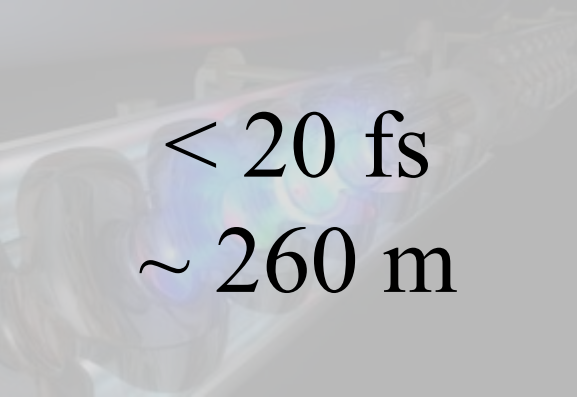
Synchrotron



10 fs (slicing)  
 $\sim 200$  m

Miao et al., *Nature*, **400**, 342 (1999)

Free Electron Laser

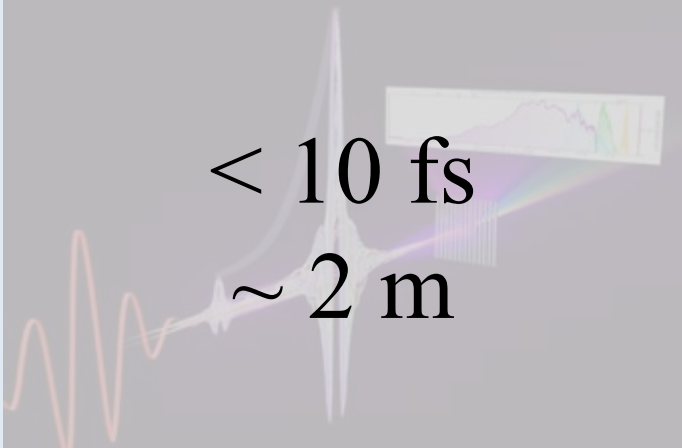


$< 20$  fs  
 $\sim 260$  m

Chapman et al., *Nature Phys.* **2**, 839 (2006)

## TABLETOP SOURCES

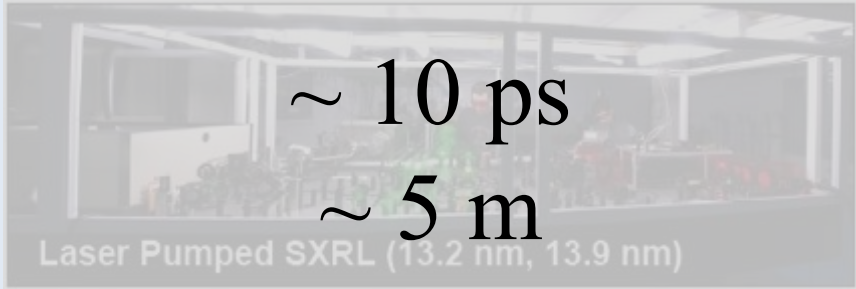
High harmonics



$< 10$  fs  
 $\sim 2$  m

T. Popmintchev et. al., *Science*, **336**, 6086, (2012)

EUV laser source at 13nm

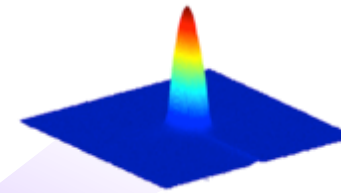
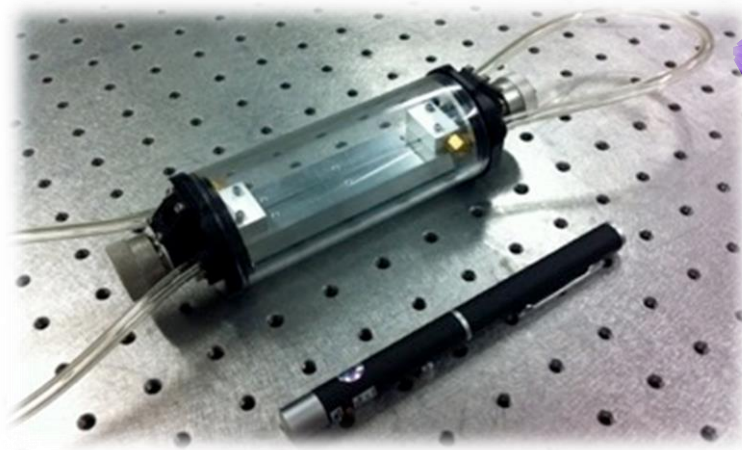
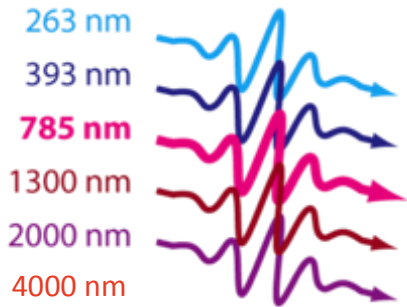


$\sim 10$  ps  
 $\sim 5$  m

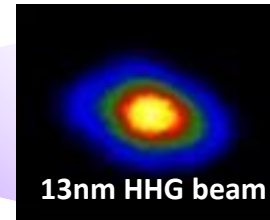
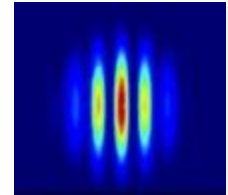
Laser Pumped SXRL (13.2 nm, 13.9 nm)

Martz et al, *Optics Letters*, **35**, 1632 (2010)

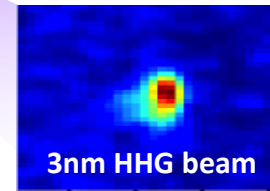
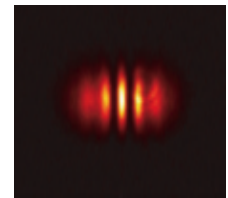
# Bright Coherent Beams From UV To keV



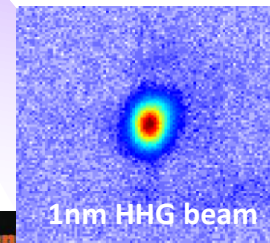
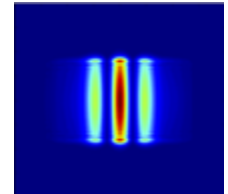
30nm HHG beam



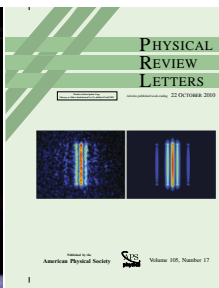
13nm HHG beam



3nm HHG beam



1nm HHG beam

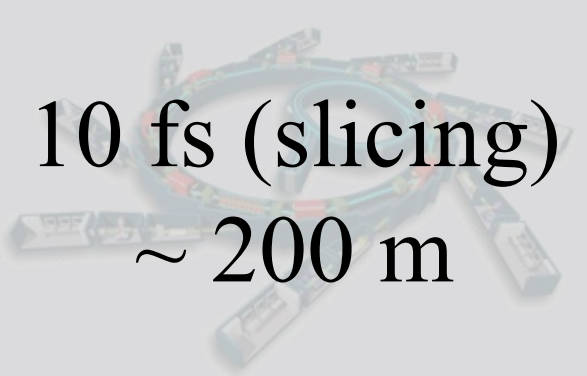


*Science* **280**, 1412 (1998)  
*Science* **297**, 376 (2002)  
*Science* **336**, 1287 (2012)  
 Pat. No. 8,462,824 (2013)

# Coherent EUV Sources

## FACILITY SCALE SOURCES

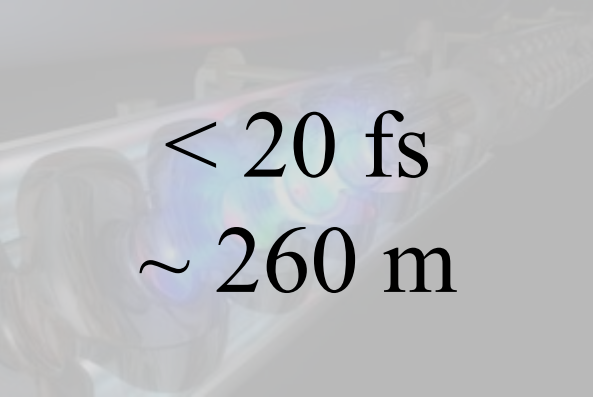
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Free Electron Laser

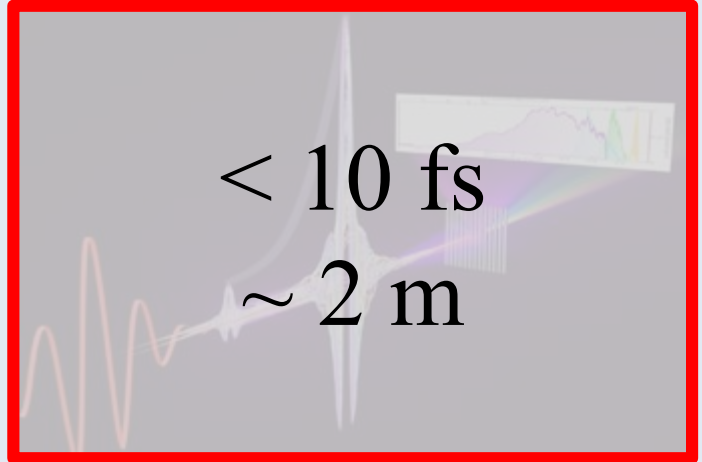


$< 20$  fs  
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Chapman et al., *Nature Phys.* **2**, 839 (2006)

## TABLETOP SOURCES

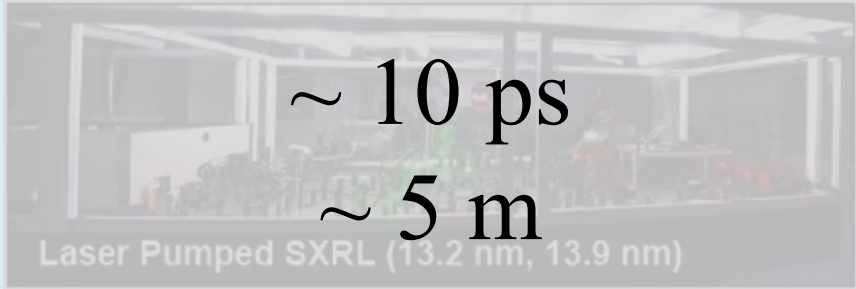
High harmonics



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T. Popmintchev et. al., *Science*, **336**, 6086, (2012)

EUV laser source at 13nm

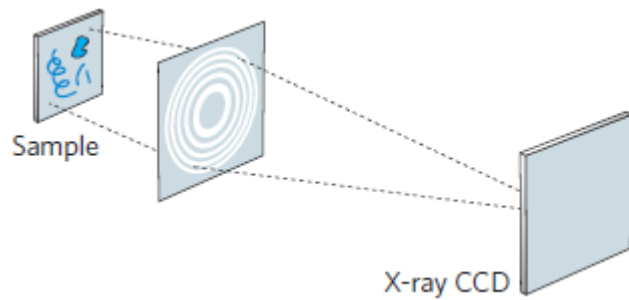


$\sim 10$  ps  
 $\sim 5$  m

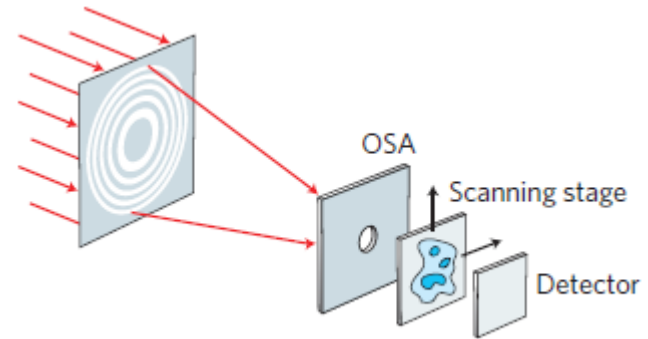
Laser Pumped SXRL (13.2 nm, 13.9 nm)

Martz et al, *Optics Letters*, **35**, 1632 (2010)

# EUV and X-ray Microscopy Techniques



Full-field Zone Plate Microscopy



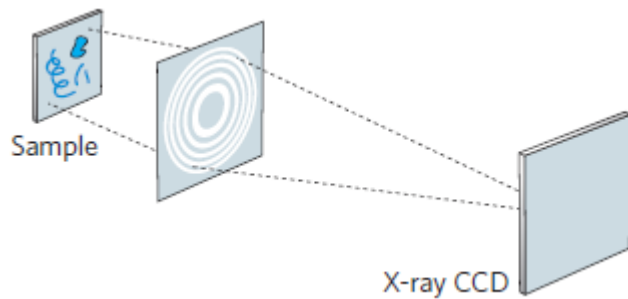
Scanning Transmission X-ray Microscopy

Sakdinawat and Attwood, *Nature Photonics*, **4**, 840 (2010)

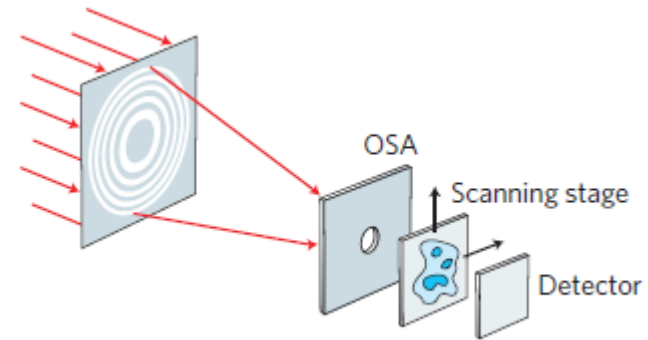
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Sakdinawat and Attwood, *Nature Photonics*, **4**, 840 (2010)

*Acta Cryst.* (1952). **5**, 843

**Some implications of a theorem due to Shannon.** By D. SAYRE, *Johnson Foundation for Medical Physics, University of Pennsylvania, Philadelphia 4, Pennsylvania, U. S. A.*

(Received 3 July 1952)

Shannon (1949), in the field of communication theory, has given the following theorem: If a function  $d(x)$  is known to vanish outside the points  $x = \pm a/2$ , then its Fourier transform  $F(X)$  is completely specified by the values which it assumes at the points  $X = 0, \pm 1/a, \pm 2/a, \dots$ . In fact, the continuous  $F(X)$  may be filled in merely by laying down the function  $\sin \pi a X / \pi a X$  at each of the above points, with weight equal to the value of  $F(X)$  at that point, and adding.

Now the electron-density function  $d(x)$  describing a single unit cell of a crystal vanishes outside the points  $x = \pm a/2$ , where  $a$  is the length of the cell. The reciprocal-lattice points are at  $X = 0, \pm 1/a, \pm 2/a, \dots$ , and hence the experimentally observable values of  $F(X)$  would suffice, by the theorem, to determine  $F(X)$  everywhere, if the phases were known. (In principle, the necessary points extend indefinitely in reciprocal space, but by using, say, Gaussian atoms both  $d(x)$  and  $F(X)$  can be effectively confined to the unit cell and the observable region, respectively.)

For centrosymmetrical structures, to be able to fill in the  $|F|^2$  function would suffice to yield the structure, for sign changes could occur only at the points where  $|F|^2$  vanishes. The structure corresponding to the  $|F|^2$  function is the Patterson of a single unit cell. This has

twice the width of the unit cell, and hence to fill in the  $|F|^2$  function would require knowledge of  $|F|^2$  at the half-integral, as well as the integral  $h$ 's. This is equivalent to a statement made by Gay (1951).

I think the conclusions which may be stated at this point are:

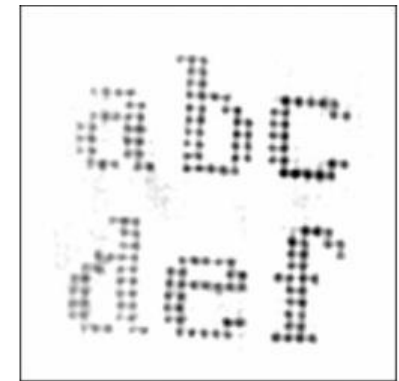
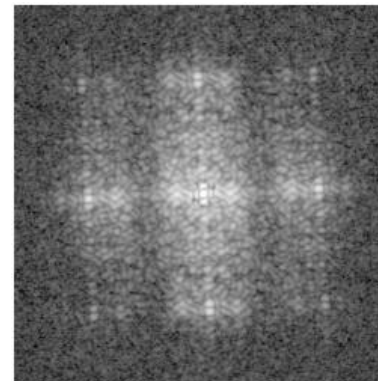
1. Direct structure determination, for centrosymmetric structures, could be accomplished as well by finding the sizes of the  $|F|^2$  at half-integral  $h$  as by the usual procedure of finding the signs of the  $F$ 's at integral  $h$ .

2. In work like that of Boyes-Watson, Davidson & Perutz (1947) on haemoglobin, where  $|F|^2$  was observed at non-integral  $h$ , it would suffice to have only the values at half-integral  $h$ .

The extension to three dimensions is obvious.

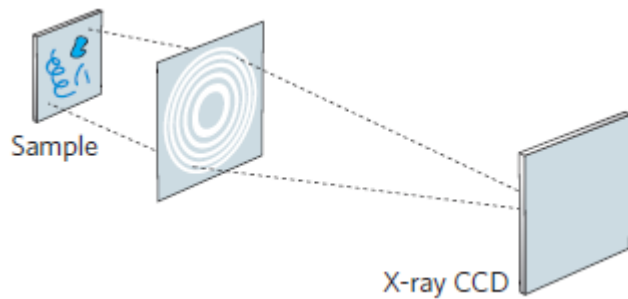
## References

- BOYES-WATSON, J., DAVIDSON, E. & PERUTZ, M. F. (1947). *Proc. Roy. Soc. A*, **191**, 83.  
GAY, R. (1951). Paper presented at the Second International Congress of Crystallography, Stockholm.  
SHANNON, C. E. (1949). *Proc. Inst. Radio Engrs.*, N.Y. **37**, 10.

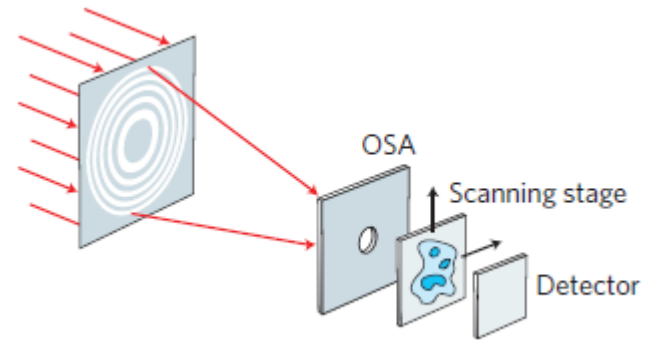


J. Miao, P. Charalambous, J. Kirz, and D. Sayre, *Nature*, vol. 400, no. 6742, pp. 342–344, Jul. 1999.

# EUV and X-ray Microscopy Techniques



Full-field Zone Plate Microscopy



Scanning Transmission X-ray Microscopy

Sakdinawat and Attwood, *Nature Photonics*, **4**, 840 (2010)

## PROSPECTS FOR LONG-WAVELENGTH X-RAY MICROSCOPY AND DIFFRACTION

D. Sayre

IBM Research Center

Yorktown Heights, New York 10598, U.S.A.

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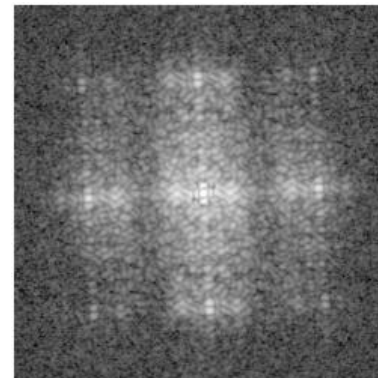
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### References

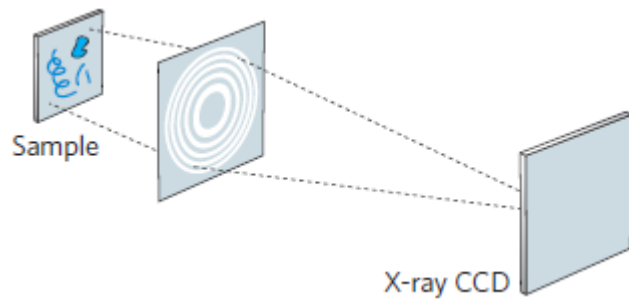
- BOYES-WATSON, J., DAVIDSON, E. & PERUTZ, M.F. (1947). *Proc. Roy. Soc. A*, **191**, 83.  
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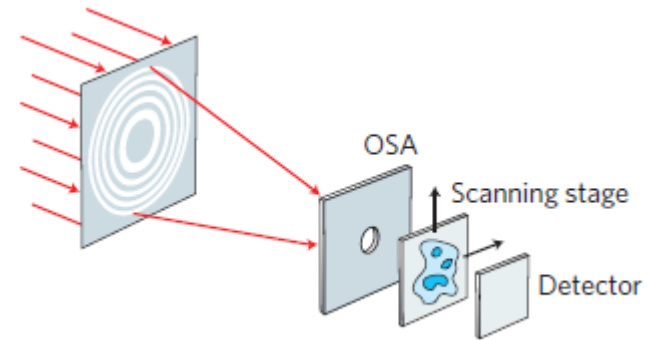
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D. Sayre, in *Lecture Notes in Physics*, Vol. 112, Imaging Processes and Coherence in Physics, 1980, pp. 229–235.

# EUV and X-ray Microscopy Techniques



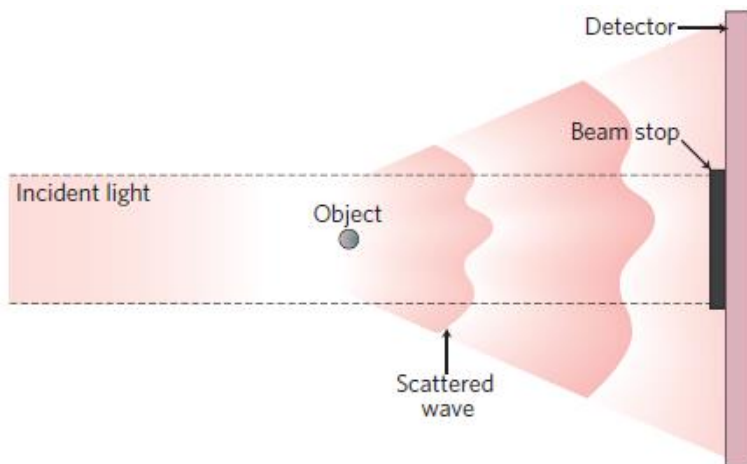
Full-field Zone Plate Microscopy



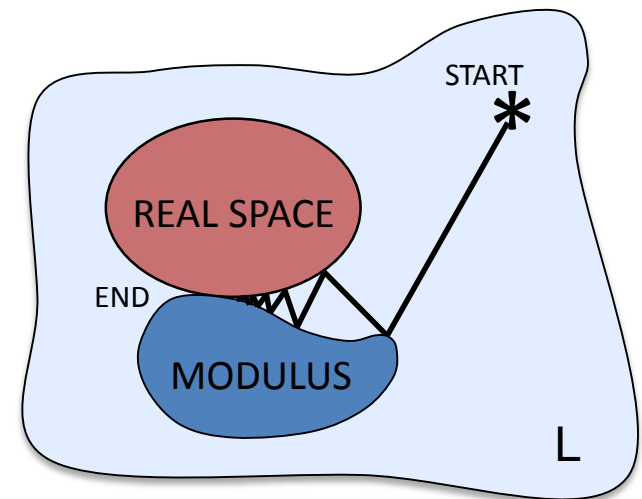
Scanning Transmission X-ray Microscopy

Sakdinawat and Attwood, *Nature Photonics*, **4**, 840 (2010)

## Coherent Diffractive Imaging (CDI)



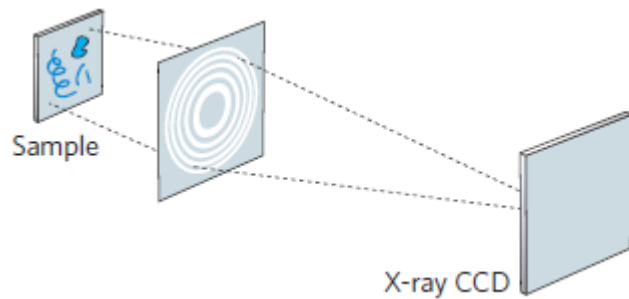
## 2D Phase Retrieval



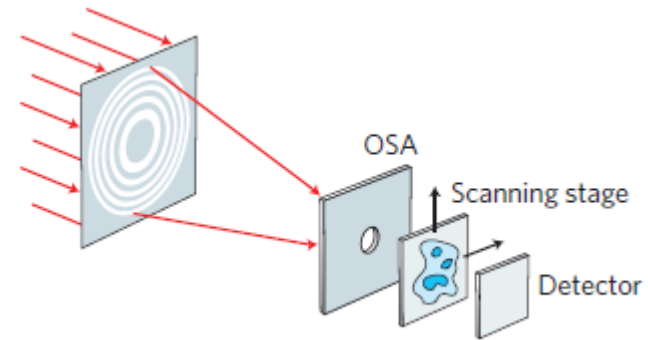
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J. R. Fienup, *Opt. Lett.*, vol. 3, no. 1, pp. 27–29, Jul. 1978.

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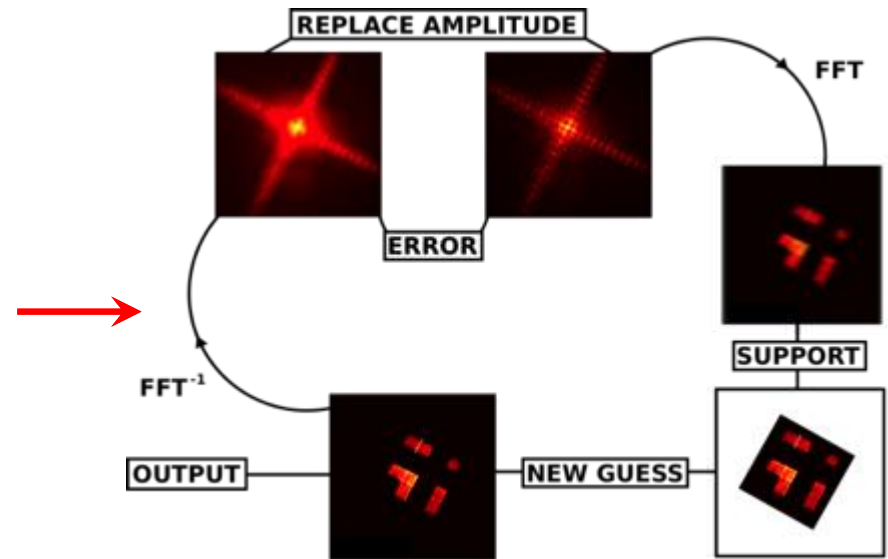
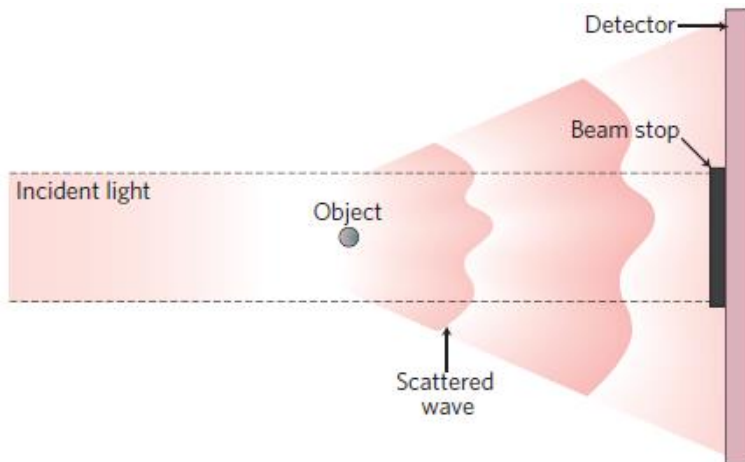
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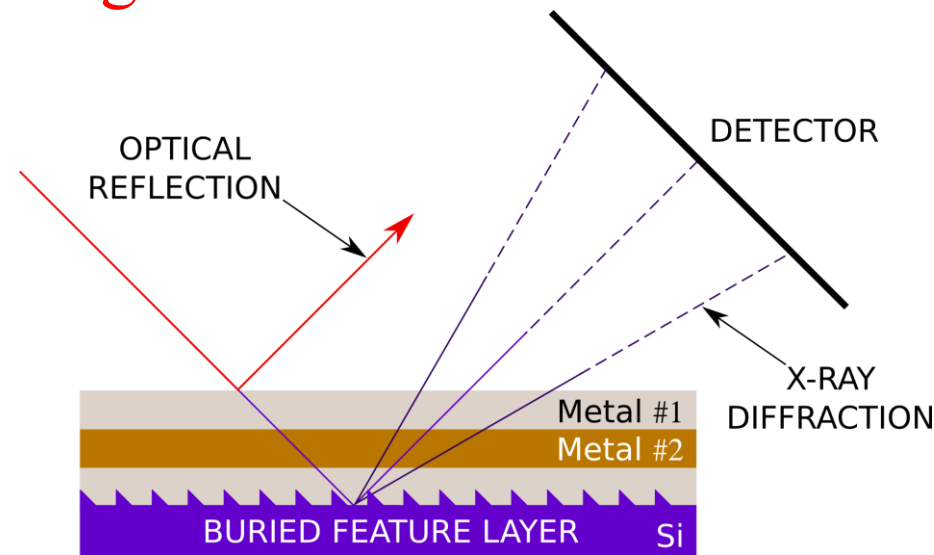
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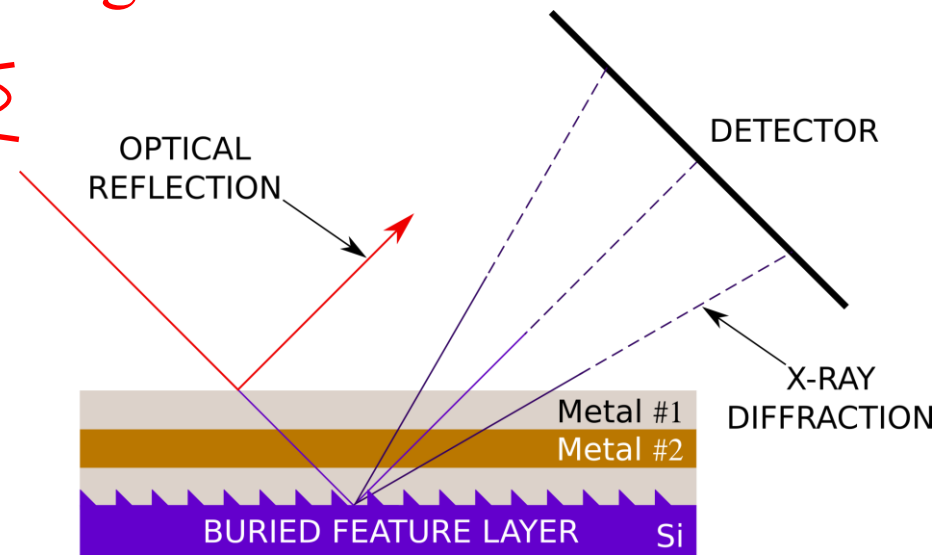
# EUV and X-ray Microscopy

- No Aberrations – **Diffraction-limited**
- Image Through Thick Samples
- **Image Buried Samples**
- Inherent Elemental Contrast Of X-rays
- Necessarily a Phase Contrast Technique
- Requires A **Coherent Beam Of Light**
- Requires An **Isolated Sample**



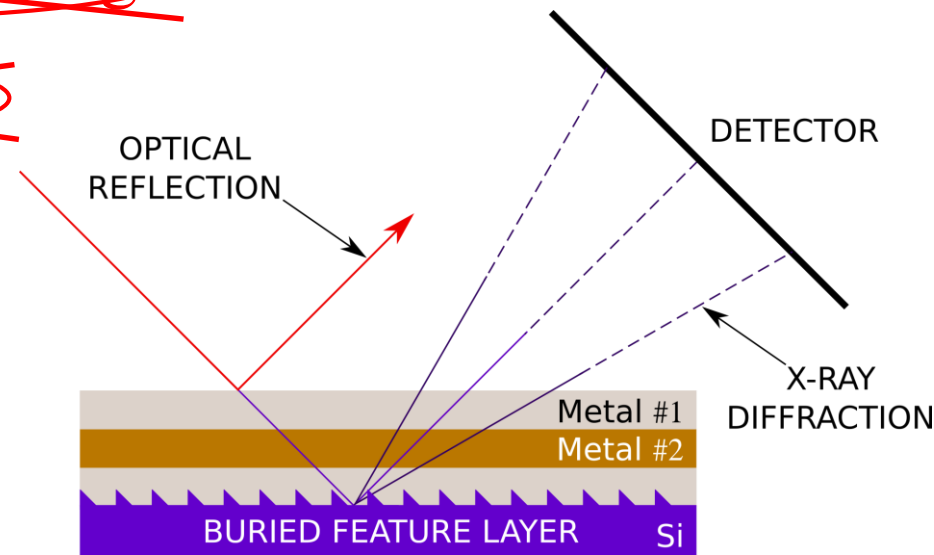
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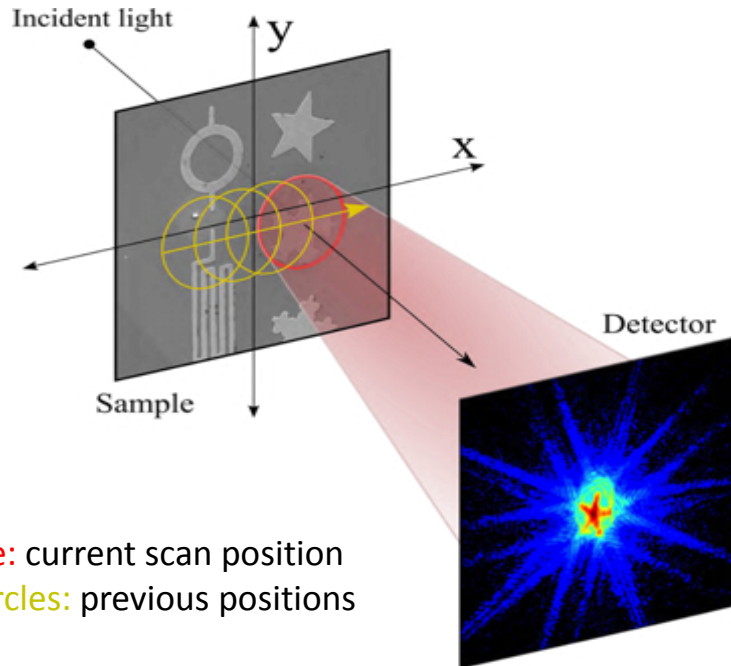


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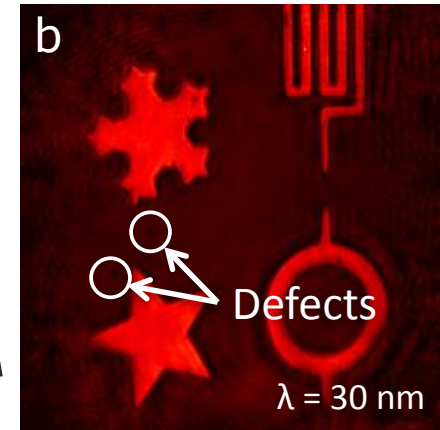


# Ptychography CDI



Red circle: current scan position  
Yellow circles: previous positions

## Phase Contrast Microscopy Through Phase Retrieval



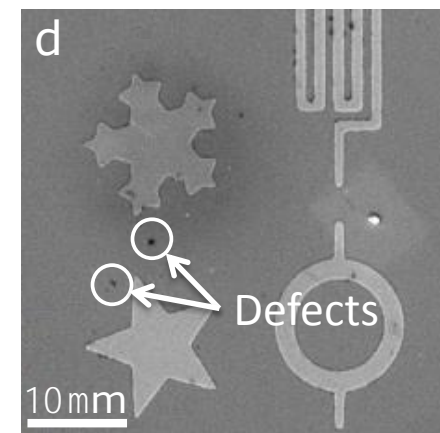
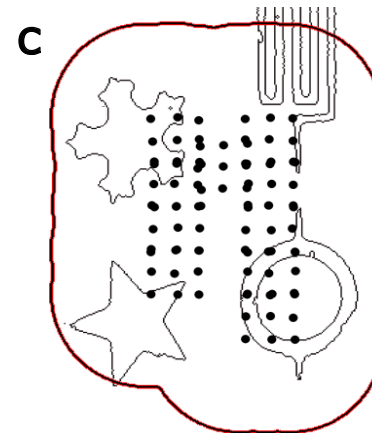
Beam scatters at many overlapping positions

### Constraints:

- 1) Measured data (many positions)
- 2) Oversampled illumination
- 3) Overlap between positions

Curved wave-front forms shadowgram

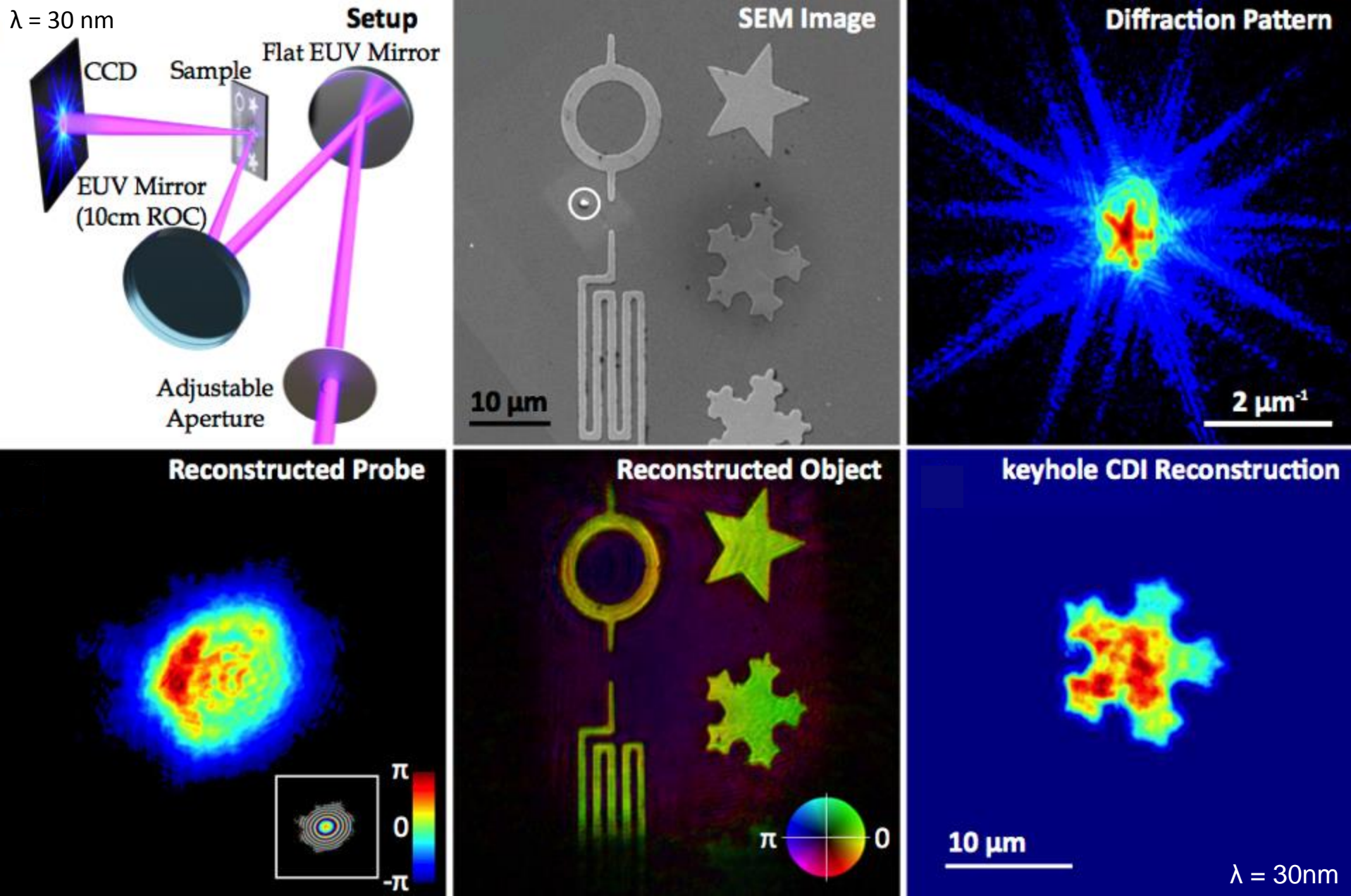
Simultaneously retrieves beam and object



Seaberg et. al, Optica 1, 1 39-44



# High Fidelity Reflection CDI with an HHG Source

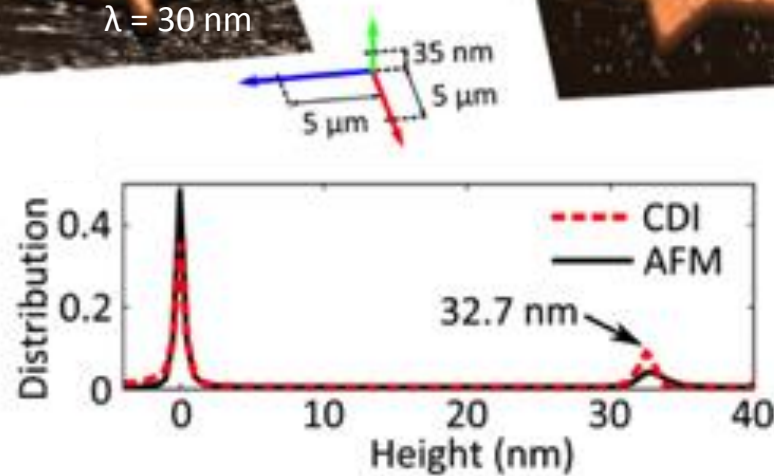


# Quantitative Topology Through Reflection CDI

CDI



AFM

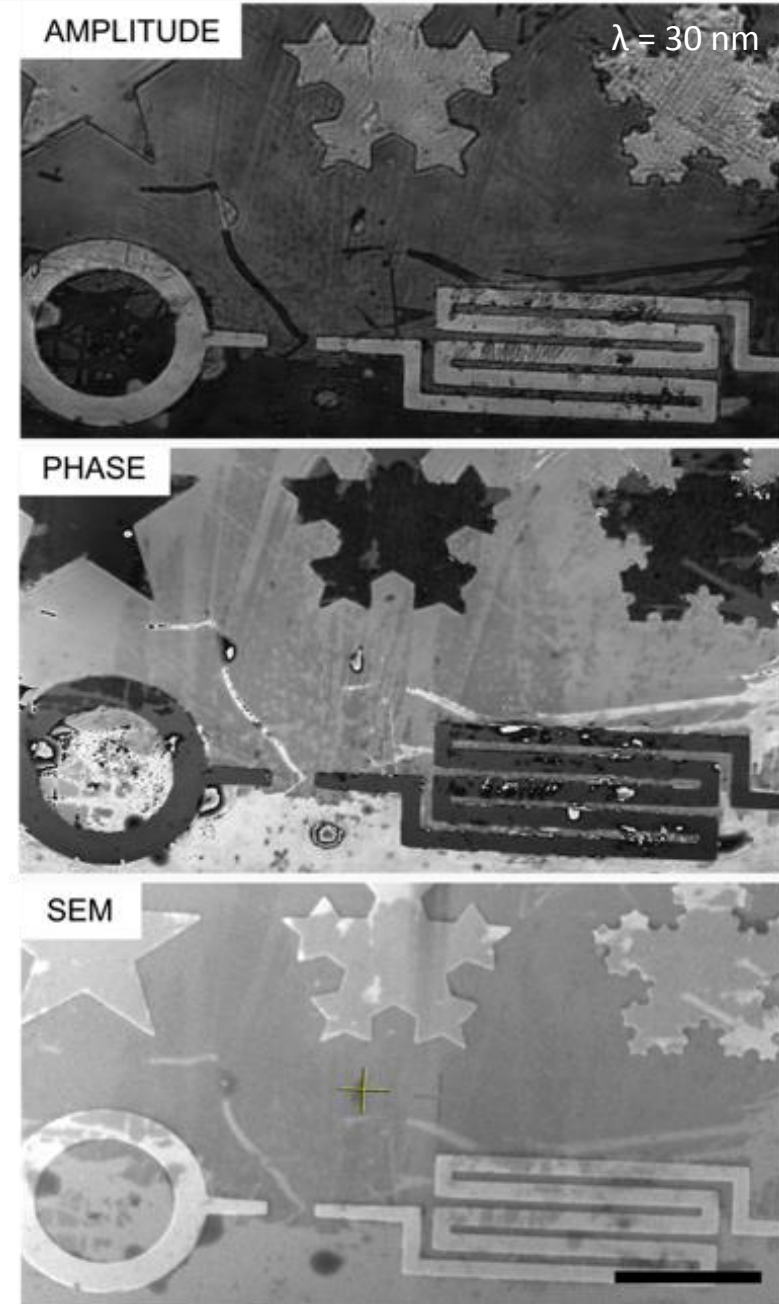
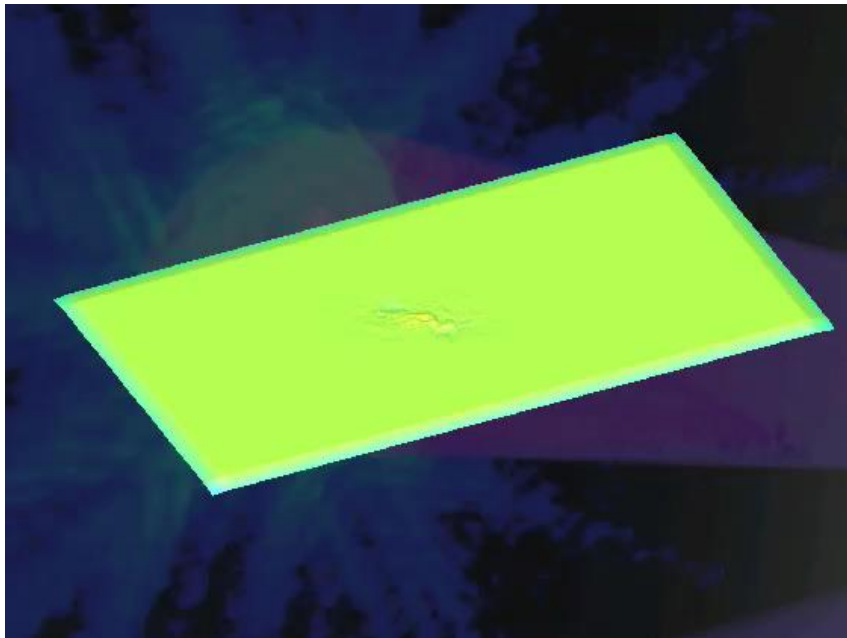


Seaberg et. al, Optica 1, 1 39-44

- Must Take Fresnel Reflections into Account (including oxide layers)
- Ellipsometry to Find oxide thickness (otherwise relative profilometry)
- CDI and AFM agree  $\pm 3\text{\AA}$

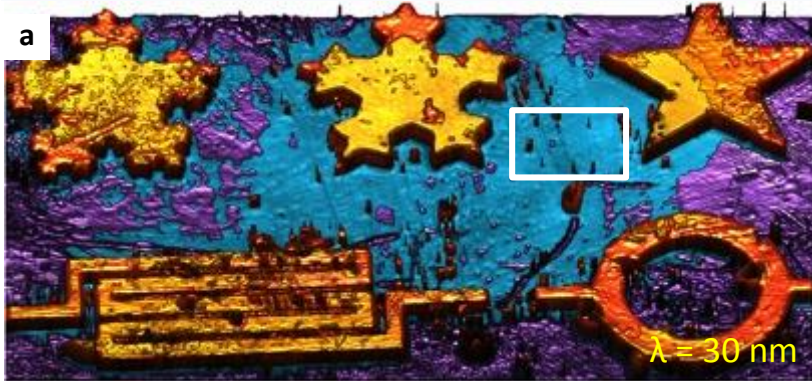
# High Fidelity Reflection CDI with an HHG Source

- 200 Positions
- 0.1 seconds/position
- 1 min total exposure
- Can be cut to 30 seconds!
- 45nm transverse resolution
- 0.5 nm axial precision
- High Contrast v. SEM
- ALGORITHMS CORRECT ERRORS





# Quantitative Topology Through Reflection CDI

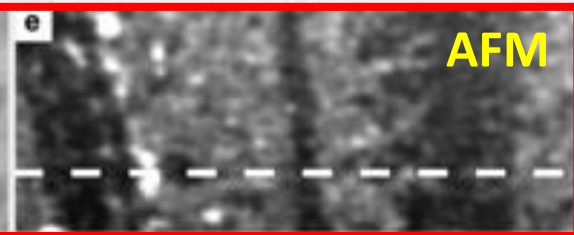
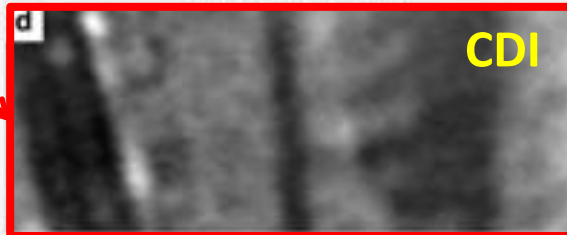
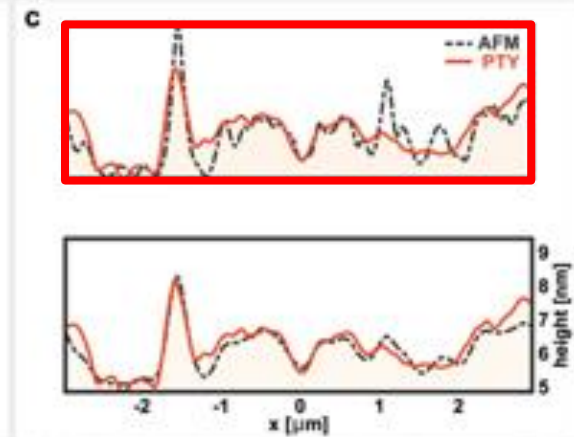
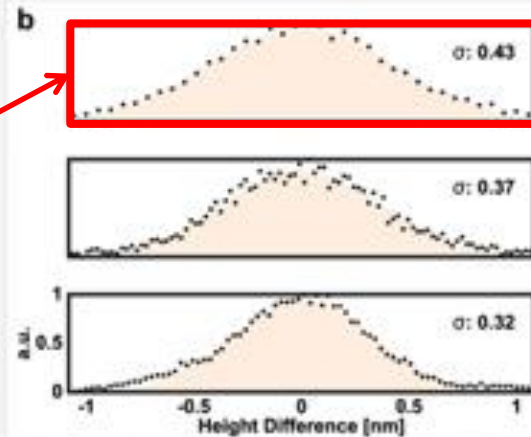


Zhang et. al, Submitted

- Small debris is weakly scattering
- Out-shined by titanium shapes
- Filter so transverse resolutions match

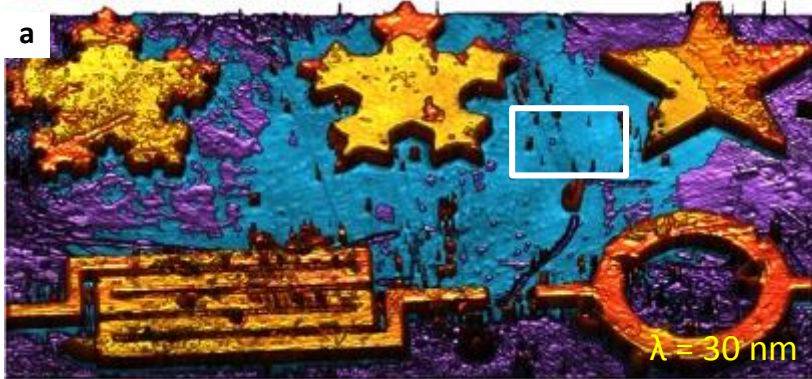
NO FILTERING

$$\sigma = 4.3 \text{ \AA}$$





# Quantitative Topology Through Reflection CDI

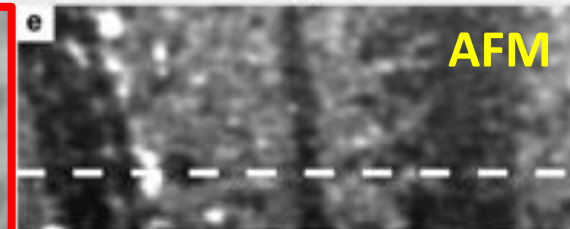
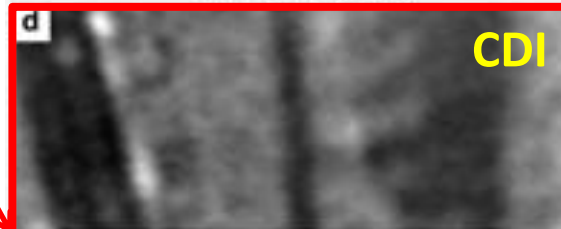
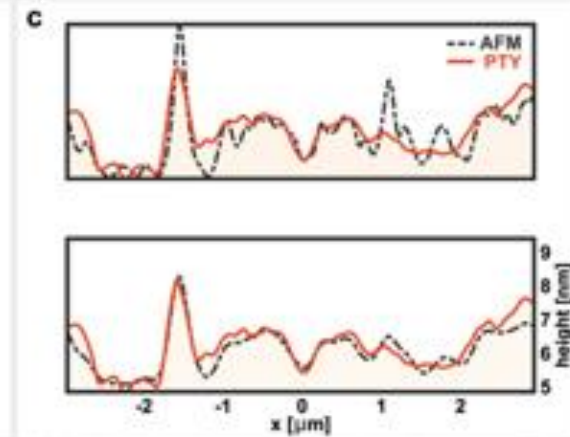
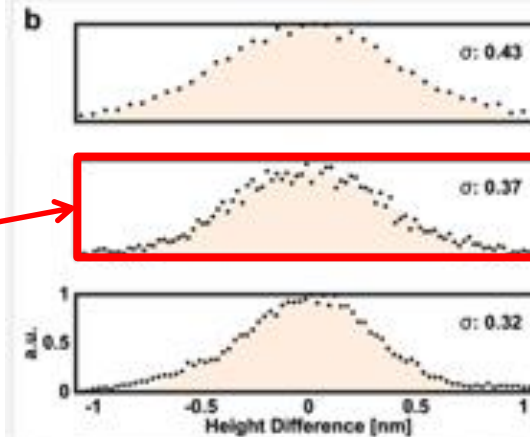


Zhang et. al, Submitted

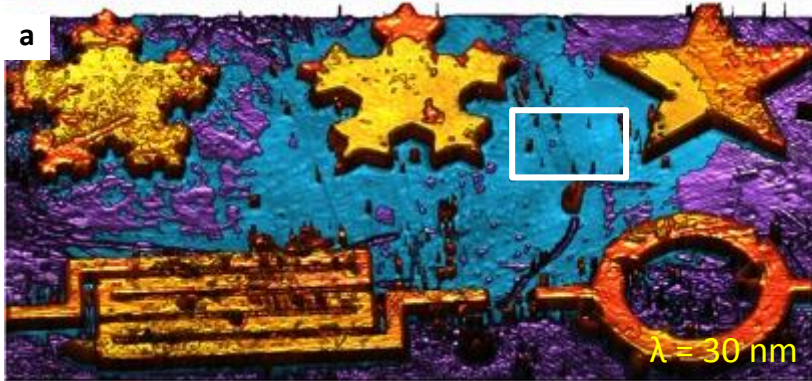
- Small debris is weakly scattering
- Out-shined by titanium shapes
- Filter so transverse resolutions match

HISTOGRAM FILTER

$$\sigma = 3.7 \text{ \AA}$$



# Quantitative Topology Through Reflection CDI

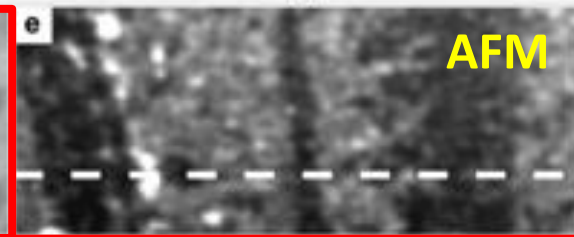
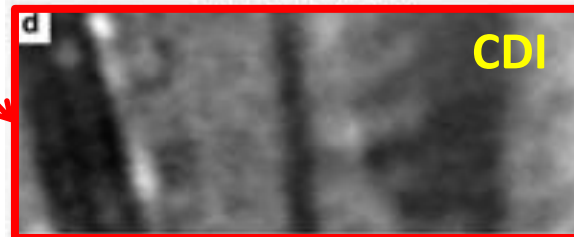
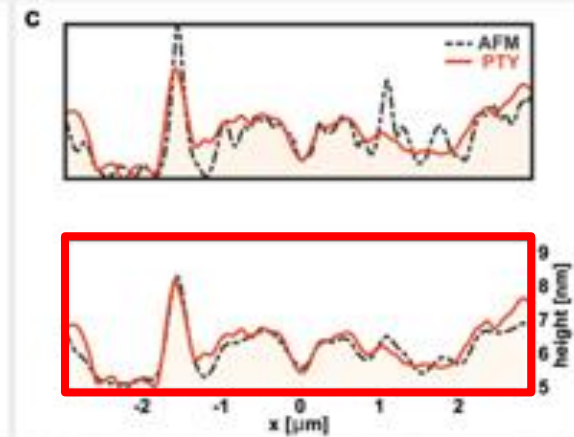
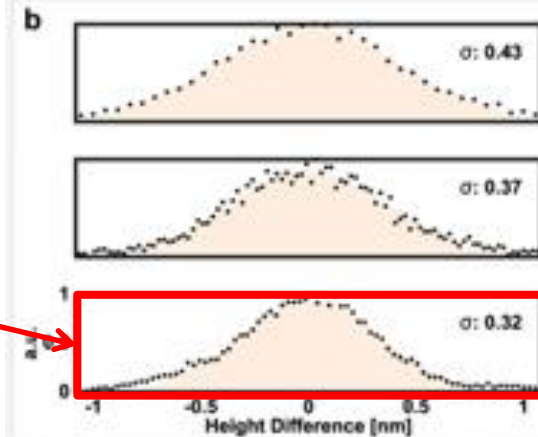


Zhang et. al, Submitted

- Small debris is weakly scattering
- Out-shined by titanium shapes
- Filter so transverse resolutions match

PSF FILTERING

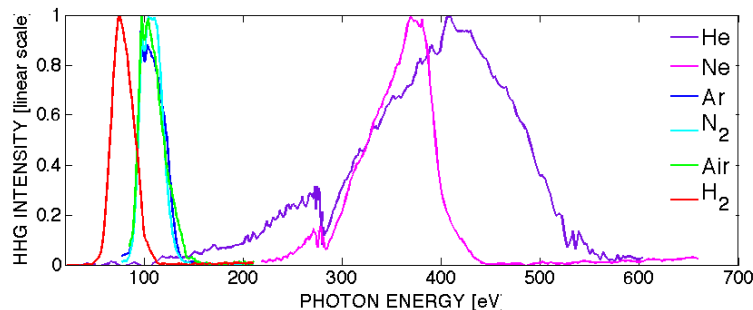
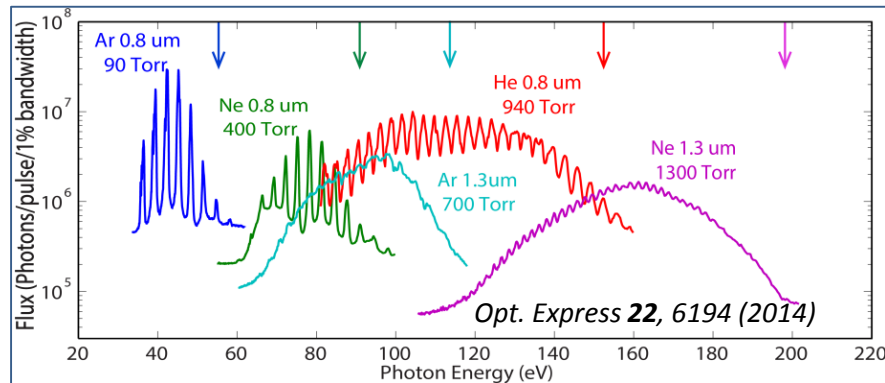
$$\sigma = 3.2 \text{ \AA}$$



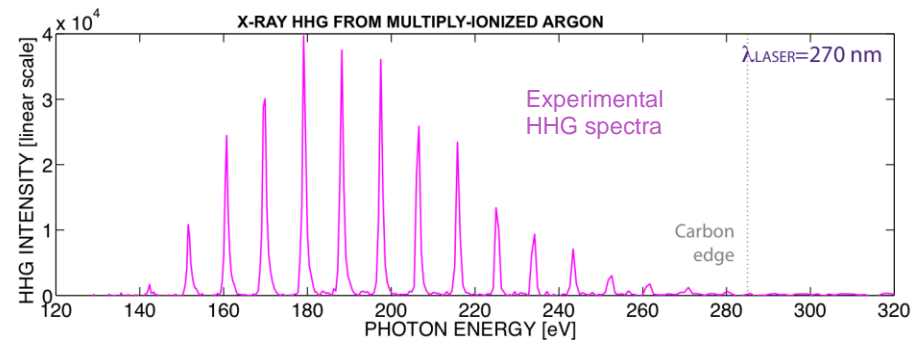
# Precise Control $\rightarrow$ High Precision Measurement

- MID-IR LASERS  $\rightarrow$  X-RAY CONTINUUM  $\rightarrow$  SPECTROSCOPY
- UV LASERS  $\rightarrow$  ISOLATED HHG PEAKS  $\rightarrow$  IMAGING/METROLOGY
- CIRCULAR POLARIZATION  $\rightarrow$  ISOLATED HHG PEAKS  $\rightarrow$  IMAGING/METROLOGY

## Mid-infrared driven soft x-ray HHG



## UV driven soft x-ray HHG

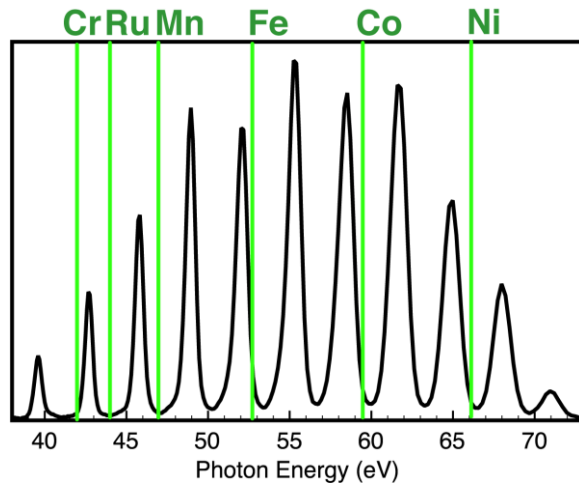


US Patent Awarded, US 61873794 (2015)  
Postdeadline paper, CLEO (2014)

# Precise Control $\longrightarrow$ High Precision Measurement

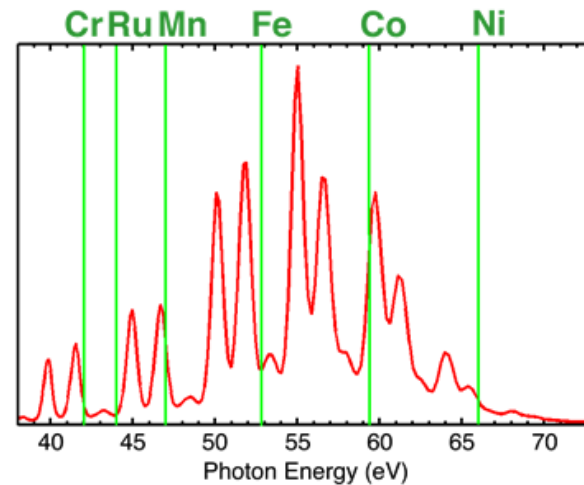
## Linearly polarized EUV HHG

- Odd harmonics of fundamental



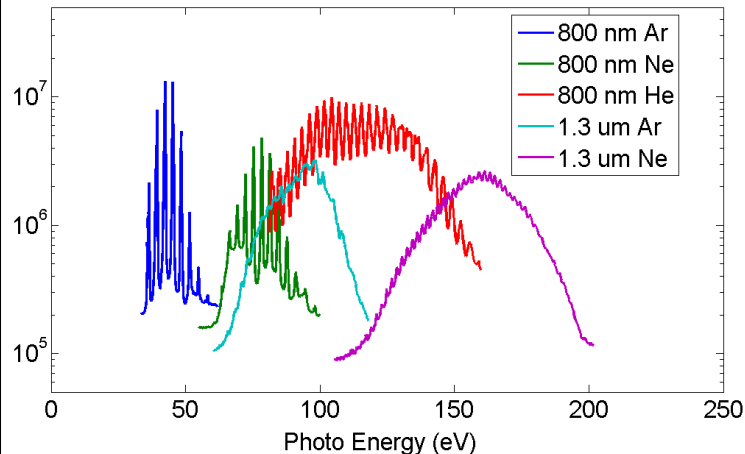
## Circularly polarized EUV HHG

- Right and left circular, every 3<sup>rd</sup> missing



## Linearly polarized soft X-ray HHG

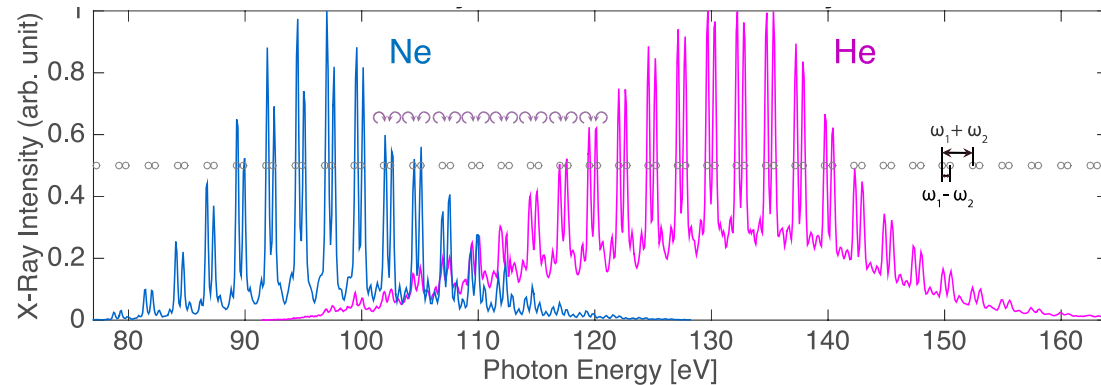
- Broad super-continuum



*Science* **336**, 1287 (2012); *Op. Ex.* **22**, 6194 (2014)

## Circularly polarized EUV HHG

- Right and left circular, narrow peaks



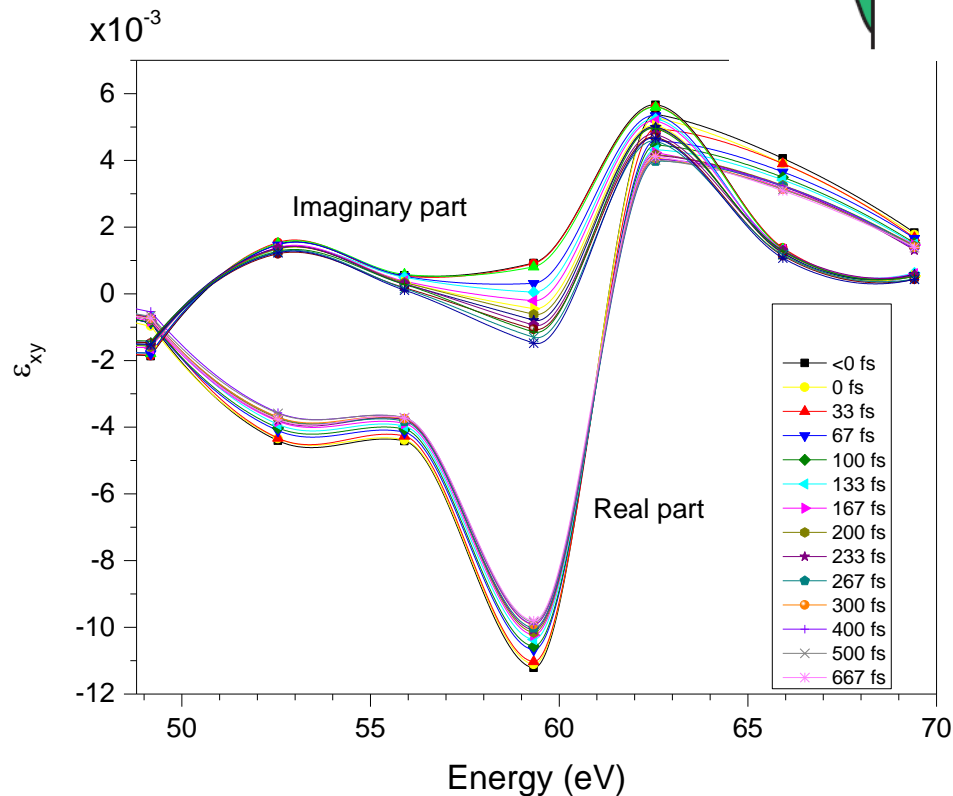
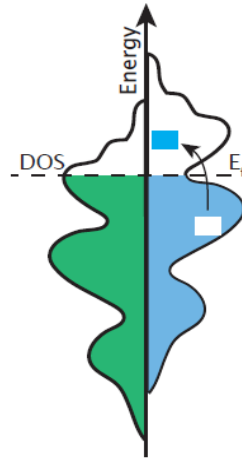
*Postdeadline paper JTh5C.1, CLEO (2015)*



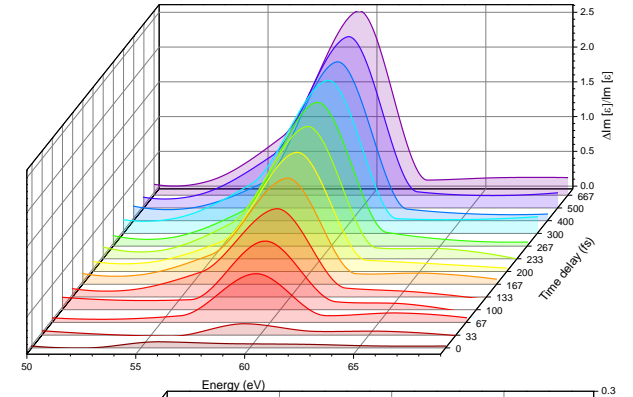
# Example: Time Resolved EUV Ellipsometry

- Extract time-dependent magneto-optic constant:  $\epsilon_{xy}(t)$  and  $\epsilon_{xx}(t)$

$$\epsilon_{xy}(\omega, t)$$

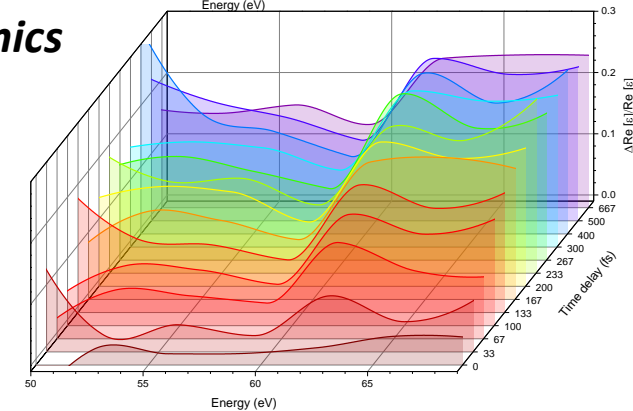


Imaginary  $\epsilon_{xy}$



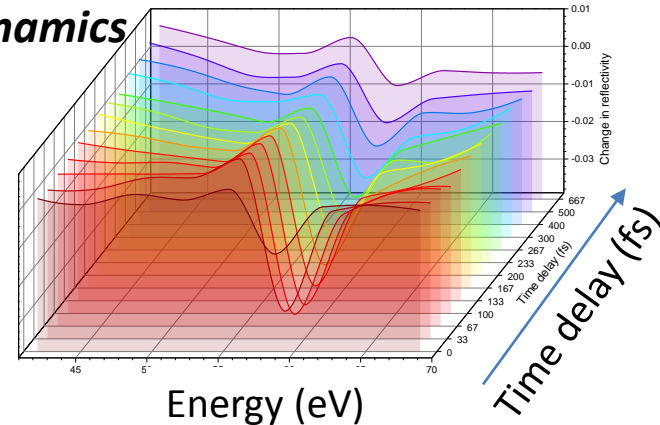
*Spin Dynamics*

Real  $\epsilon_{xy}$

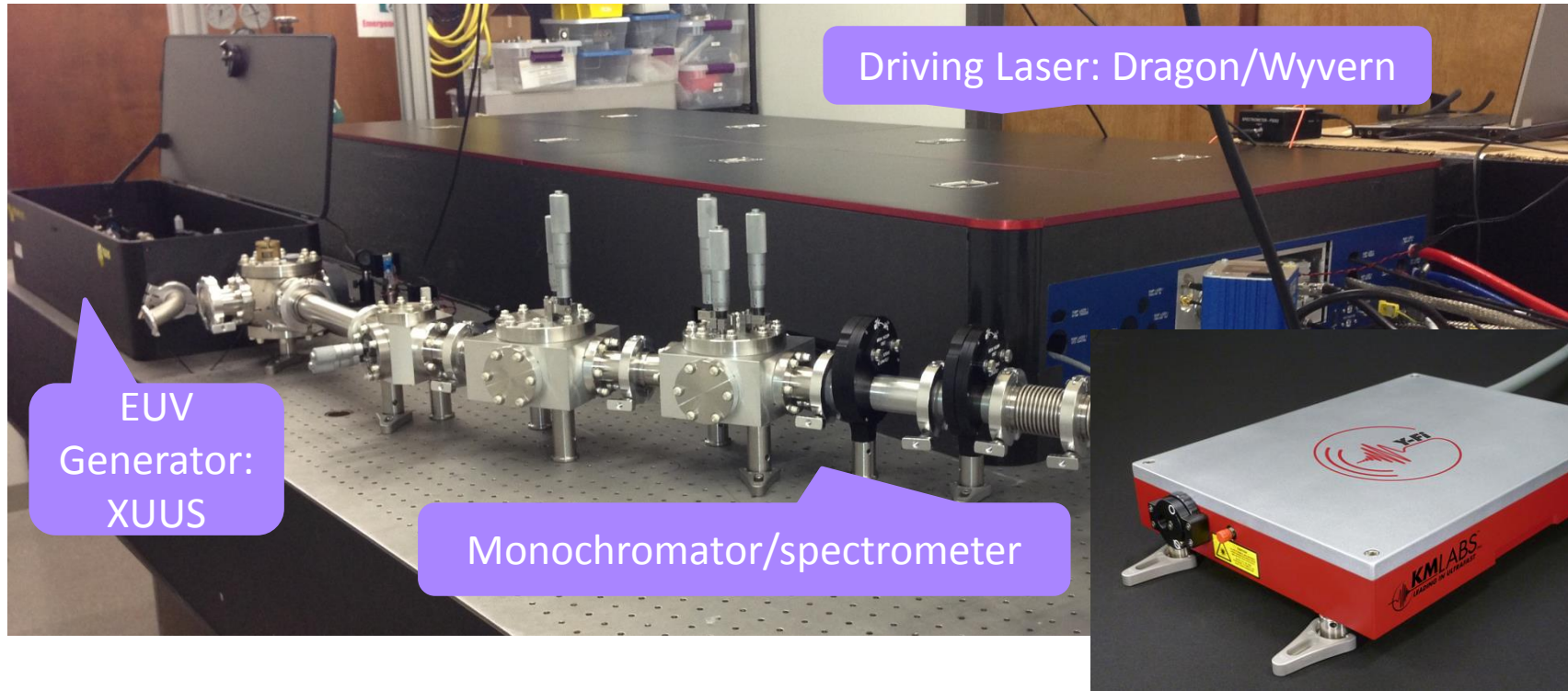


*Charge Dynamics*

Reflectivity



# Commercial HHG Source and Beam-line



- Interest from biomedical industry allowing KMLabs to engineer laser – **Y-Fi**
- Phase IIb DOE SBIR grant allowing us to **engineer** commercial EUV source with ultrastable automatic laser alignment - **XUUS 4**





# XUUS at CLEO

**XUUS4**  
generation coherent  
EUV source

EUV output  $>10^{12}$  ph/sec  
Stable, efficient HHG waveguide  
Active, 4-axis pointing stabilization  
Growing family of beamline modules

Even coherent EUV  
tool for a wide range  
of applications

*Laser Focus World*  
*Science*

**KMLABS**  
LEADING IN ULTRAFAST

**XUUS4**

Industry Leading  
High Harmonic  
Generation

**20 KMLABS**  
LEADING IN ULTRAFAST

*femtosecond* **1994-2014** *EUV* *X-Ray*

**XUUS**

**RED DRAGON**

**KMLABS**  
LEADING IN ULTRAFAST

**WYVERN**  
PRISM AWARDS  
2013 WINNER

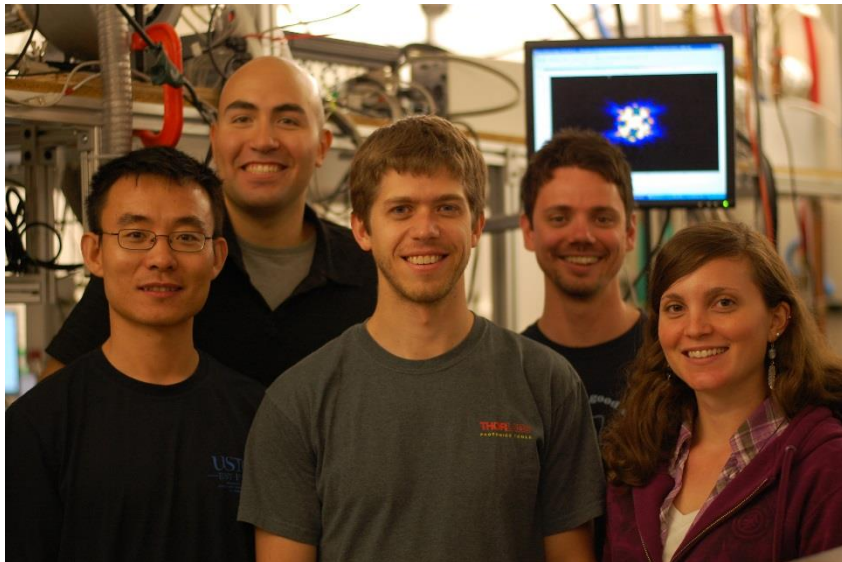
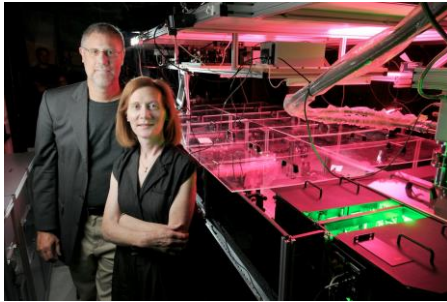
**RED DRAGON**

**SMART** TABLE



# Acknowledgements

- **Funding Agencies:** DARPA, SRC, NSF, AFOSR
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- **Students:** Matthew Seaberg, Bosheng Zhang, Dennis Gardner, Elisabeth Shanblatt, Christina Porter, Robert Karl, Michael Tanksalva, Charlie Bevis
- **Collaborators:** Jianwei Miao, Eric Gullikson, Carmen Menoni, Bill Schlotter, Erik Anderson, Brian Abbey...

